



## DECLARATION

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TO: [illegible]

I, IKUO FUJINO, a Japanese Patent Attorney registered No. 9188, of Okabe International Patent Office at No. 602, Fuji Bldg., 2-3, Marunouchi 3-chome, Chiyoda-ku, Tokyo, Japan, hereby declare that I have a thorough knowledge of Japanese and English languages, and that the attached pages contain a correct translation into English of the priority documents of Japanese Patent Application No. 10-313938 filed on October 19, 1998 in the name of CANON KABUSHIKI KAISHA.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made, are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this *27th* day of September, 2002

IKUO FUJINO



Corres. and Mail.  
**BOX AF**

AF/1745

**COMMUNICATION UNDER 37 C.F.R. § 1.116  
EXPEDITED PROSECUTION - GROUP ART UNIT 1745**

03500.013929

PATENT APPLICATION

A14  
9.30.20

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

TOMONARI HORIKIRI ET AL.

Application No.: 09/417,832

Filed: October 14, 1999

For: GEL ELECTROLYTE, CELL  
AND ELECTROCHROMIC  
ELEMENT

)  
:  
) Examiner: M. Ruthkosky

)  
:  
) Group Art Unit: 1745

)  
:  
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:  
) September 26, 2002

Commissioner for Patents  
**Box: AF**  
Washington, D.C. 20231

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**LETTER TRANSMITTING  
SWORN TRANSLATION OF PRIORITY APPLICATION**

Sir:

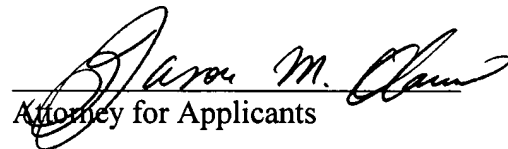
Further to the Amendment After Final Rejection in the above-captioned application, which was filed on September 23, 2002, Applicants submit herewith a sworn translation of Japanese Patent Application No. 10-313938, filed October 19, 1998, from which the subject application claims priority under 35 U.S.C. § 119. See 37 C.F.R. § 1.55; see also M.P.E.P. § 201.15.

The filing of this sworn translation removes JP 11-185836 (JP '836), which was published on July 9, 1999, as prior art as to the claims supported by the Japanese application.

Entry of this sworn translation is respectfully requested.

Applicants' undersigned attorney may be reached in our New York office by telephone at (212) 218-2100. All correspondence should continue to be directed to our below listed address.

Respectfully submitted,

  
Attorney for Applicants

Registration No. 48,512

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PATENT OFFICE  
JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy  
of the following application as filed with this Office.

Date of Application: October 19, 1998

Application Number: Japanese Patent Application  
No. 10-313938

Applicant(s): CANON KABUSHIKI KAISHA

November 12, 1999

Commissioner; TAKAHIKO KONDO  
Patent Office

(seal)

Certificate No.11-3078859

10-313938

[Name of the Document]	Patent Application
[Reference No.]	3682089
[Date]	October 19, 1998
[Address to]	Commissioner of Patent Office TAKESHI ISAYAMA
[International Classification]	H01M 6/22
[Title of the Invention]	GEL ELECTROLYTE
[Number of Claims]	5
[Inventor]	
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[Inventor]	
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[Attorney]

[Identification No.]

100069017

[Name]

WATANABE TOKUHIRO

[Indication of Official Fee]

[Way of Payment]

Prepaid

[Prepayment Ledger No.]

015417

[Amount]

21000

[List of Filed Materials]

[Material]

Specification

1

[Material]

Drawing

1

[Material]

Abstract

1

[General Power of Attorney]

9703886

[Name of Document] SPECIFICATION

[Title of the Invention] GEL ELECTROLYTE

[CLAIMS]

1. A gel electrolyte containing at least a  
5 gelling agent and a material of high ion conductivity  
being liquid at working temperature.

2. The gel electrolyte of claim 1, wherein said  
material of high ion conductivity is a salt being  
10 liquid at room temperature.

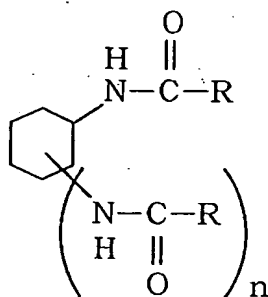
3. The gel electrolyte of claim 1, wherein said  
gelling agent is a self-assembling compound which gels  
forming a polymer associated body by the aid of an  
15 intermolecular force, such as hydrogen bonding,  
coordination bonding and the like.

4. The gel electrolyte of claim 3, wherein said  
self-assembling compound has at least one group, as the  
20 substituent showing capability of hydrogen bonding,  
selected from the group consisting of carbamate, amide,  
urea, carboxyl, alkoxy, hydroxyl, and phosphate groups.

5. The gel electrolyte of claim 3 or 4, wherein  
25 said self-assembling compound is selected from the  
group consisting of the compounds represented by the  
following formulae (1) to (16):

[Che1]

5

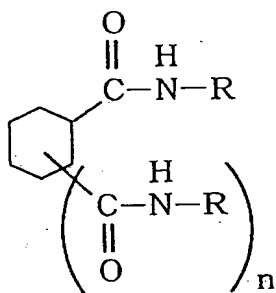


( 1 )

( n = 0, 1, 2 )

[Che2]

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( 2 )

( n = 0, 1, 2 )

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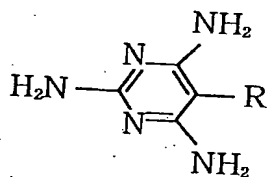
[Che3]



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[Che4]

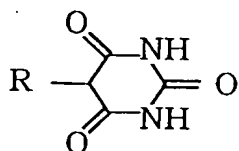
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[Che5]

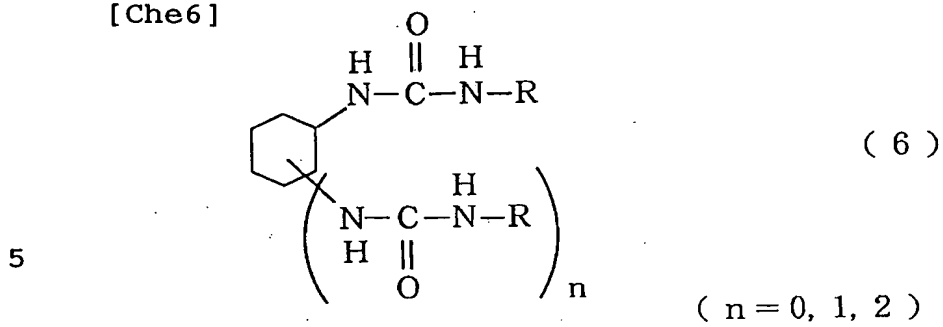
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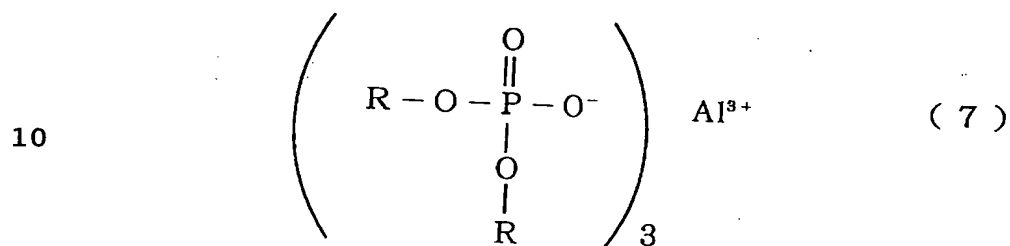
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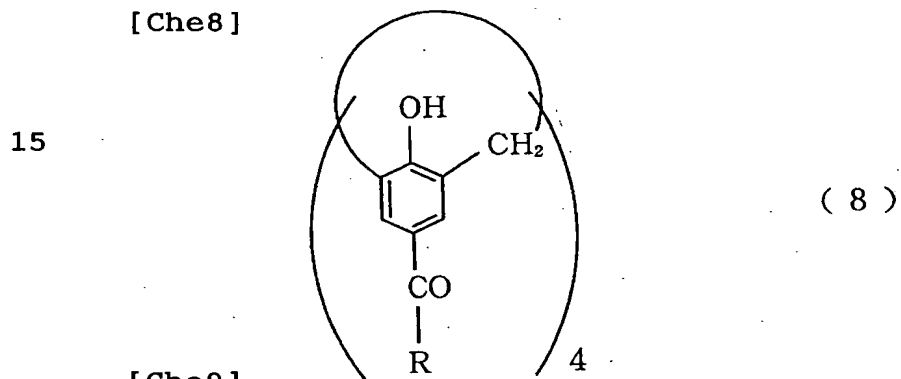
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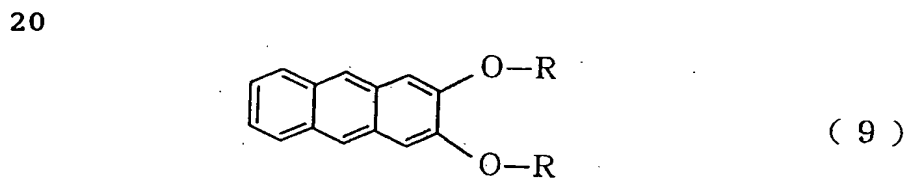
[Che7]



[Che8]



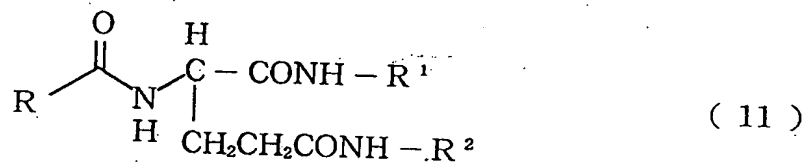
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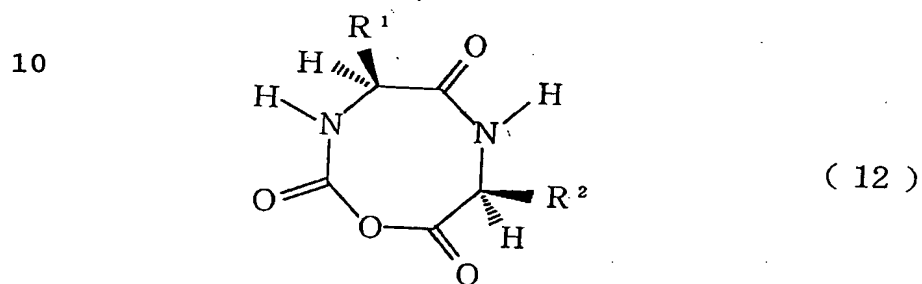
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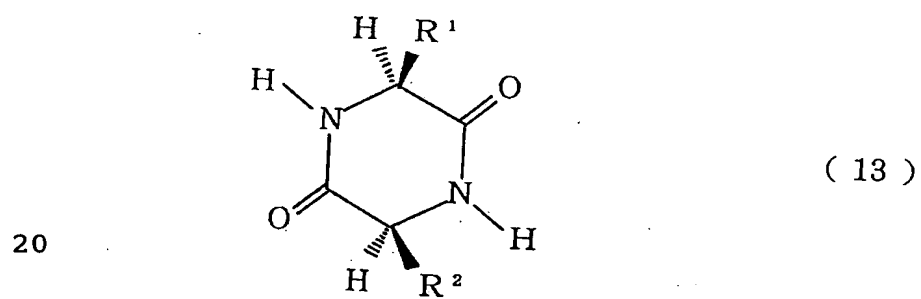
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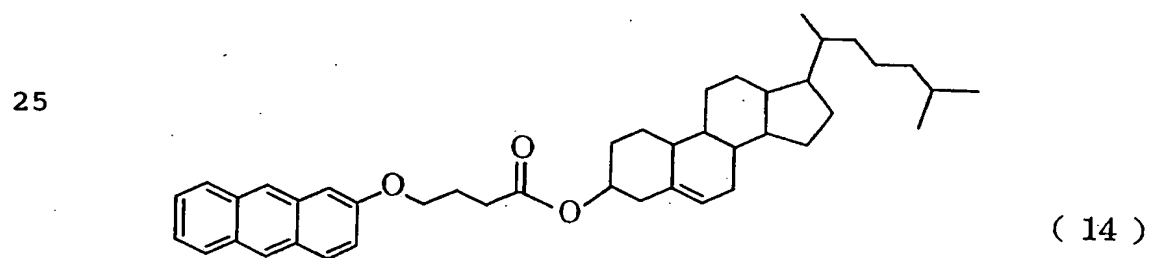
[Che12]



[Che13]

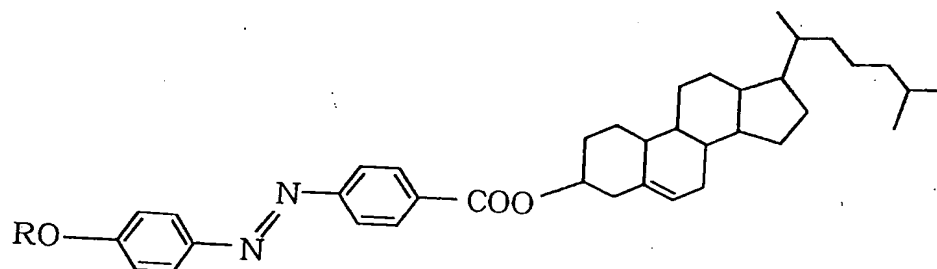


[Che14]



[Che15]

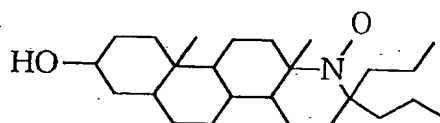
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( 15 )

[Che16]

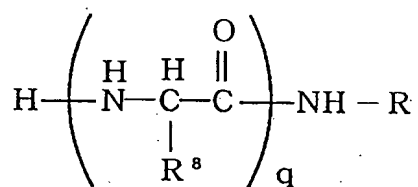
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( 16 )

[Che17]

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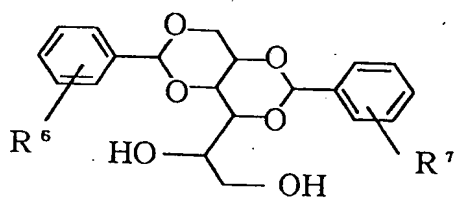


( 17 )

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[Che18]

25



( 18 )

wherein, R, R<sup>1</sup> and R<sub>2</sub> are each hydrogen, or a straight-chain or branched aliphatic hydrocarbon group having a carbon number of 1 to 29; R<sup>3</sup> is an amino acid monomer or dimer with a protected amino group; R<sup>4</sup> is an aliphatic hydrocarbon having a carbon number of 1 to 29 or an aryl group; R<sup>5</sup> is a straight-chain aliphatic group having a carbon number of 1 to 29 and being substituted with one hydroxyl group; R<sup>6</sup> and R<sup>7</sup> are each an aliphatic hydrocarbon group having a carbon number of 1 to 29 or an aryl group; R<sup>8</sup> is hydrogen, or an aliphatic hydrocarbon group having a carbon number of 1 to 5 or aryl group; n is 0, 1 or 2; q is an integer of 2 to 20.

[Detailed Explanation of the Invention]

[0001]

[Technical field of the invention]

5 This invention relates to a gel electrolyte, which  
can be used as a solid thin film electrolyte useful for  
electrochemical elements such as cells, condensers,  
electrochemical sensors and electrochromic elements.

[0002]

[Related Background Art]

10 Recently, importance of electrolyte has been  
increasing with the technical progress in various  
electrochemical elements such as cells and  
electrochromic elements. In many conventional  
electrochemical elements, liquid electrolytes with  
15 supporting electrolytes dissolved in water or organic  
solvents. However, the liquid electrolyte has various  
disadvantages, such as the leakage due to temporal  
deterioration in long term storage or due to damage of  
the electrochemical element, and difficulty in making  
20 the electrochemical element compact in size or into a  
thin film. Therefore, the electrolyte solidification  
has been actively studied to develop solid thin-film  
electrolytes easy to handle, high in safety and in the  
ion transport number.

25 [0003]

Since inorganic materials such as alumina are low  
in ion conductivity at ordinary temperature, polymers

have been studied mainly for the solid electrolytes.

[0004]

Researches on polymer-based solid electrolytes fall into two general approaches.

- 5           One is a gel electrolyte holding a liquid electrolyte in a polymer matrix network.

[0005]

- Such a gel electrolyte, swelled polymer with a liquid electrolyte, can be produced by polymerizing the monomer in a liquid electrolyte solution, or by  
10           immersing a polymer matrix that has been polymerized beforehand in a liquid electrolyte for swelling.

[0006]

- In such a gel electrolyte, the polymer matrix  
15           basically functions to include the liquid electrolyte with no contribution to ion conductance, and the liquid electrolyte in the polymer matrix is mainly responsible for ion conductance. This type of gel electrolyte has a high ion conductivity, close to that of the liquid  
20           electrolyte, because of the comparatively free movement of the ions in the polymer matrix.

[0007]

- The other approach is the polymer-based solid electrolyte where a supporting electrolyte is dissolved  
25           in a polymer having a polyether structure, such as polyethylene oxide, polypropylene oxide, a derivative or copolymer thereof or the like. The polyether-

structured polymer can dissolve one monovalent cation by 4 oxygen atoms of the ether structure, and the cations hop the ether structure in the polymer chain for conductance. This type of electrolytes basically  
5 contains no solution, and is essentially free of leakage problems.

[0008]

[Problems to be solved by the invention]

However, such conventional polymer solid  
10 electrolytes need a large quantity of polyether, and as a result, the gel electrolytes obtained tend to be much lower in conductivity than the original electrolytic solution itself.

[0009]

15 It is known that a compound having self-assembling characteristics (self-assembling compound), which forms a fibrous associated body driven by the intermolecular force such as hydrogen bonding, can gel a liquid with a very small quantity. Since such a self-assembling  
20 compound becomes a gel as the fibrous associated bodies entwine with each other to form a network structure, to loose fluidity, and holding a liquid in its voids, the gel is expected to have flexible and fine functions as a material differing from conventional 3-dimensionally  
25 cross-linked structure of polymers, or a random hydrogen bond net work structure of a natural polymer such as agar and gelatin.

[0010]

A thin film of organic electrolyte of a high ion transport number can be formed with such a self assembling compound, since it can gel with a small amount of a liquid and the gelled substance (associated bodies) is larger than the polymer chain. Moreover, by using a liquid electrolyte of which dissolution in a solvent is not necessary, prevention of conductivity lowering can be expected.

10 [0011]

Heretofore, a gel electrolyte formed from a self-assembling compound and a liquid electrolyte such as a salt liquid at room temperature has been disclosed in Japanese Patent No. 2599763 where dibenzylidene sorbitol derivative is used. However, dibenzylidene sorbitol derivatives are relatively unstable, freeing aldehydes by the action of temperature, moisture or the like to give off offensive odor and, it may color in some cases. Thus, the addition of a stabilizer, such as sorbic acid, potassium sorbate, an alkali metal compound, alkaline organic amine or the like is required to stabilize the gel over a long period of time (Japanese Patent Publication Nos. H7-17648 and H5-202055).

25

[0012]

In view of the aforementioned technical



background, the present invention intends to provide a stable gel electrolyte of which conductivity is prevented from deterioration by using a gelling agent capable of gelling a liquid electrolyte with a small amount, without color change.

[0013]

[Means to solve the problem]

Accordingly, the present invention provides a gel electrolyte containing at least a gelling agent and a material of high ion-conductivity which is liquid at working temperature.

[0014]

The liquid material of high ion-conductivity is preferably a salt being liquid at room temperature. The gelling agent is a self-assembling compound having at least one substituent selected from the group consisting of carbamate, amide, urea, carboxyl, alkoxy, hydroxy and phosphate groups, which gels forming a polymeric associated body by intermolecular interactions, e.g., hydrogen or coordination bonding..

[0015]

The gel electrolyte of the present invention comprises a gelling agent and a liquid substance of high ionic conductivity which is liquid at working temperature.

In the present invention, used as a gelling agent is a self-assembling compound which gels forming a

polymeric associated body by intermolecular interactions, e.g., hydrogen or coordination bonding.

[0016]

The self-assembling compounds useful as the gelling agent for the present invention preferably have at least one substituent selected from the group consisting of carbamate, amide, urea, carboxyl, alkoxy, hydroxy and phosphate groups. It is preferable to use at least one compound selected from the compounds represented by the formulas (1) to (18).

[0017]

The compounds represented by formulas (1) to (18) are described below.

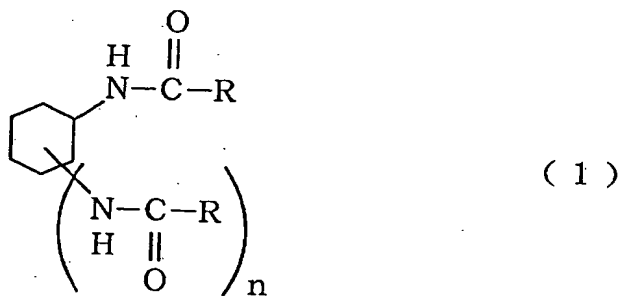
[0018]

Compounds Nos. (1) and (2)

[0019]

[Che19]

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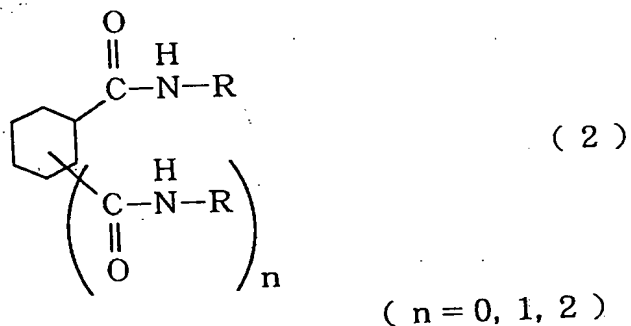


[0020]

( n = 0, 1, 2 )

[Che20]

25



[0021]

The cyclohexane derivatives useful for the present invention, represented by formulae (1) and (2) fall into two general categories; one having an amide substituent bonded to the cyclohexane ring through a nitrogen atom and the other having an amide substituent bonded to the ring through a carbon atom, both substituents capable of hydrogen bonding. Number of the amide groups and their positions on the cyclohexane ring are not defined. The cyclohexane ring may have substituents not participating in hydrogen bonding, e.g., methyl or ethyl.

[0022]

The substituent represented by R in formula (1) or (2) is hydrogen, or a C<sub>1</sub> - C<sub>29</sub> straight-chain or branched aliphatic hydrocarbon group. More specifically, the straight-chain aliphatic hydrocarbon substituents useful for the present invention include nonyl, decyl, undecyl, lauryl, tridecyl, myristyl, pentadecyl, palmityl, heptadecyl, stearyl, arachidyl, docosanoyl, tricosyl, tetracosyl, hexacosyl and triaconsyl groups, and branched aliphatic hydrocarbon substituents include 3,5,5-trimethylhexyl, 2-hexyldecyl and 2-methylhexadecyl groups, but not limited to the above. These groups represented by R may be the same or different.

[0023]

The substituent may have the same structure as the liquid electrolyte.

(n) in formula (1) or (2) is an integer of 0, 1 or 2.

5 [0024]

Compound No. (3)

[0025]

[Che21]

$R^3-NH-R^4$  (3)

10 [0026]

The amino acid derivatives represented by formula (3) may be used, where  $R^3$  is an amino acid monomer or dimer having protected amino group(s), and  $R^4$  is a  $C_1 - C_{29}$  aliphatic hydrocarbon or an aryl group. The substituent for protecting the amino group may be those normally used for peptide synthesis including carbobenzoxy group (Z group) and quaternary butyloxycarbonyl group (Boc group).

[0027]

20 The amino acid may any known amino acid, and the dimer may be composed of the same or different amino acids. They are preferably optically active. More specifically, the preferable amino acids include Z-L-valyl, Z-D-valyl, Z-L-valyl-L-valyl, Z-L-isoleucyl, Z-D-valyl-L-valyl, Z-L-leucyl- $\beta$ -alanyl, Z-L-valyl-L-leucyl, and Z-L-valyl- $\beta$ -alanyl.  $-NHR^4$  is octadecyl amino (excluding  $NHC_{18}H_{35}$ ), nonylamino, decylamino,

25

undecylamino, laurylamino, tridecylamino,  
tetradecylamino, pentadecylamino, hexadecylamino,  
heptadecylamino, stearyl amino, or nonanodecacylamino  
group, but not limited to the above.

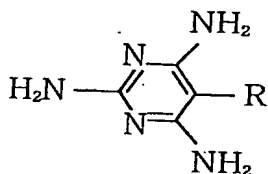
5 ]0028]

Compounds Nos. (4) and (5)

[0029]

[Che22]

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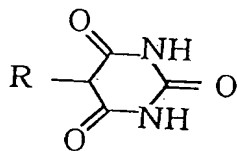


( 4 )

[0030]

[Che23]

15



( 5 )

[0031]

20 The triaminopyrimidine derivative represented by  
formula (4) and the barbiturate derivative represented  
by formula (5) may be used to form a gel by mixing them  
each other. R in formula (4) or (5) is, as defined  
above, a straight-chain or branched aliphatic  
hydrocarbon group having a carbon number of 1 to 29.  
25 The preferable compounds include dodecyl, hexadecyl and  
3,7-dimethyl octyl groups, but not limited to them.

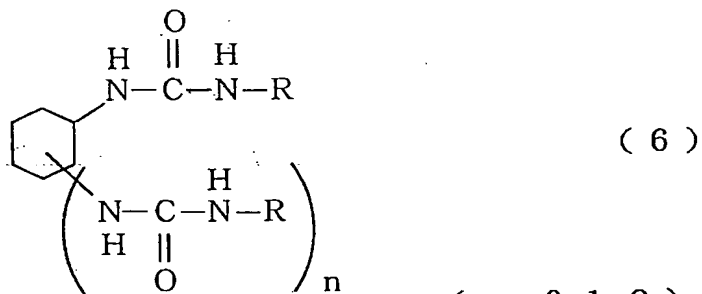
[0032]

Compound No. (6)

[0033]

[Che24]

5



10

[0034]

( n = 0, 1, 2 )

The alkyl urea derivatives represented by formula (6) are compounds having at least one urea group.

R is as defined above, but not specifically limited. These substituents may be the same or different each other.

15

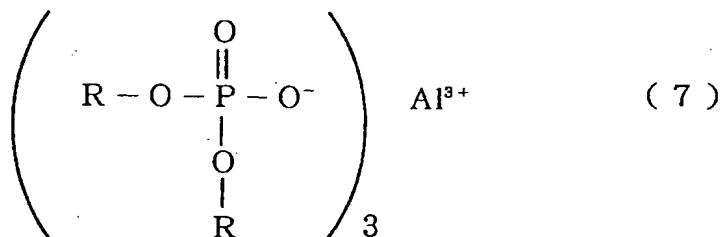
[0035]

Compound No. (7)

[0036]

[Che25]

20



[0037]

25

The aluminum phosphate derivatives represented by formula (7) can be also used, where R is as defined above, but not specifically limited. The two

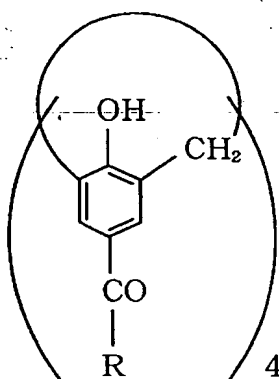
substituents in the derivative may be the same or different.

[0038]

[Che26]

5

10



( 8 )

[0040]

15

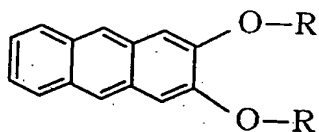
The phenol-based cyclic-oligomer derivatives represented by formula (8) can be used, where R is as defined above, preferably undecyl, but not specifically limited.

[0041]

20

Compound No. (9)

[Che27]



( 9 )

25

[0043]

The dialkoxanthracene derivatives represented by formula (9) can be used, where R is as defined above,

preferably decyl or hexadecyl, but not specifically limited.

[0044]

Compound No. (10)

5 [0045]

[Che28]



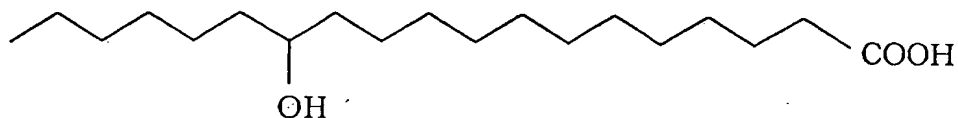
[0046]

10 Formula (10) represents hydroxycarboxylic acid derivatives represented by the following 12-hydroxyoctadecanoic acid.

[0047]

[Che29]

15



[0048]

20 In the formula  $R^6$  is a straight-chain aliphatic hydrocarbon group having a carbon number of 1 to 29, substituted by one hydroxyl group. Examples of these compounds also 3-hydroxypropyl acid, 2-hydroxybutyric acid, 3-hydroxymyristic acid and 16-hydroxyhexadecanoic acid.

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[0049]

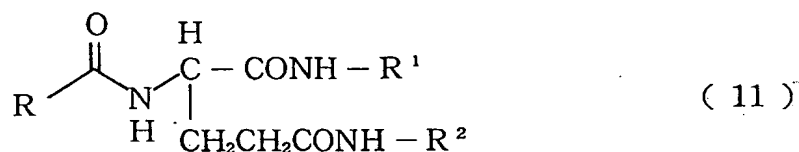
Compound No. (11)



[0050]

[Che30]

5



[0051]

Representative compounds of the carbonylamino acid diamide derivatives are carbonylglutamic acid bisamide derivatives represented by formula (11), wherein, R, R<sup>1</sup> and R<sup>2</sup> are each hydrogen, or a straight-chain or branched aliphatic hydrocarbon group having a carbon number of 1 to 29. More specifically, the straight-chain aliphatic hydrocarbon groups include nonyl, decyl, undecyl, lauryl, tridecyl, myristyl, pentadecyl, palmityl, heptadecyl, stearyl, arachidyl, docosanoyl, tricosyl, tetracosyl, hexacosyl and triaconsyl groups, and branched aliphatic hydrocarbon substituents including 3,5,5-trimethylhexyl, 2-hexyldecyl and 2-methylhexadecyl groups, but not limited thereto. The amino acid is also not limited.

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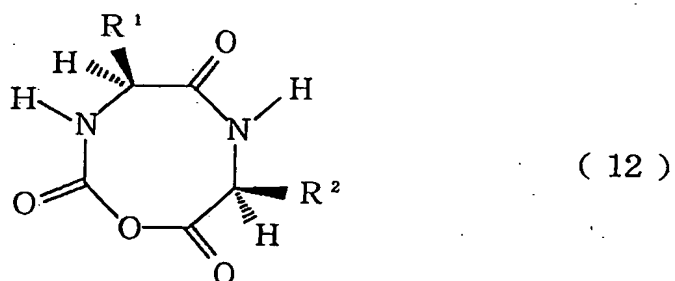
[0052]

Compound No. (12)

[0053]

[Che31]

25



[0054]

The cyclic depsipeptide derivatives represented by formula (12) may be used, wherein  $R^1$  and  $R^2$  are as defined above, but not specifically limited.

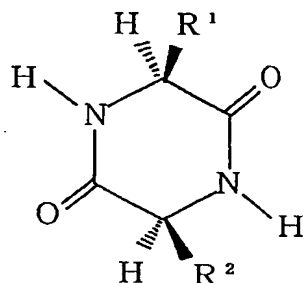
5 [0055]

Compound No. (13)

[0056]

[Che32]

10



( 13 )

[0057]

15 The cyclic dipeptide derivatives represented by formula (13) is obtained by condensation reaction using a neutral and an acidic amino acid as the starting materials. The starting amino acids are not specifically limited.  $R^1$  and  $R^2$  in the formula (13) are as defined above, but not specifically limited.

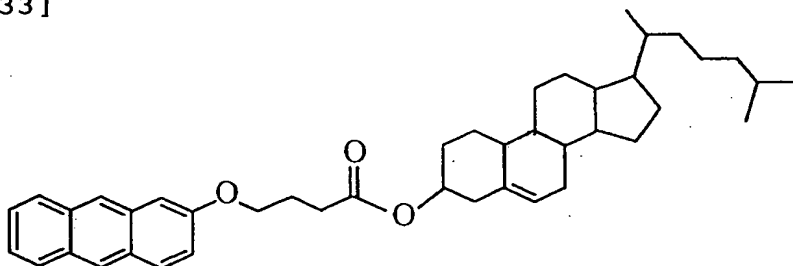
20 [0057]

Compound No. (14)

[0059]

[Che33]

25



( 14 )

[0060]

The cholesterol derivatives represented by formula (14) can be used.

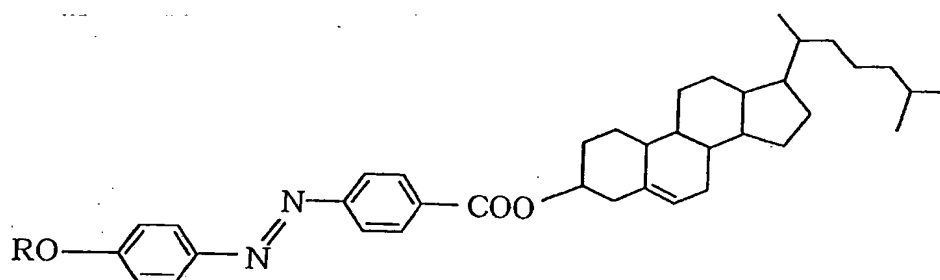
[0061]

5 Compound No. (15)

[0062]

[Che34]

10



( 15 )

[0063]

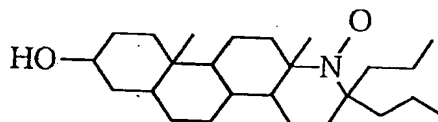
The cholesterol derivatives represented by formula (15) can be used, where R is as defined above, but not specifically limited.

[0064]

Compound No. (16)

[0065]

20 [Che35]



( 16 )

[0066]

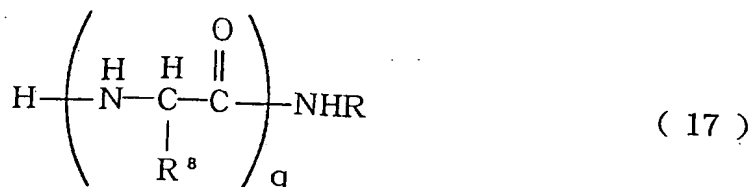
25 The spin-labeled steroid derivatives represented by formula (16) can be used.

[0067]

Compound No. (17)

[Che36]

5



[0068]

The oligo ( $\alpha$ -amino acid) derivatives represented by formula (17) has an  $\alpha$ -amino acid oligomer skeleton, where R is hydrogen or an aliphatic hydrocarbon group having a carbon number of 1 to 29; R<sup>8</sup> is hydrogen, or a C<sub>1</sub> - C<sub>5</sub> aliphatic hydrocarbon or an aryl group; and (q) is 2 to 20.

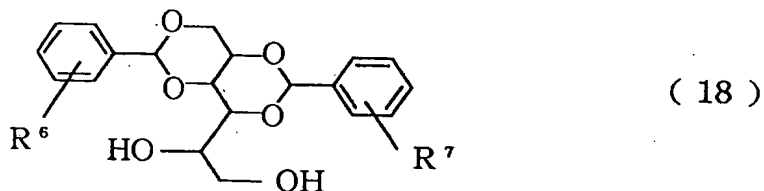
Degree of polymerization (q) is preferably 5, 10 or 20, and R<sup>9</sup> is preferably a long-chain ester group, but not specifically limited.

[0069]

Compound No. (18)

[0070]

20 [Che37]



25 [0071]

The dibenzylidene sorbitol derivative represented by formula (18) can be used, wherein R<sup>6</sup> and R<sup>7</sup> are each

Table 1: Examples of liquid electrolytes useful for the present invention

Types	Concrete examples
<p>5 Imidazolium salts</p>	<div data-bbox="706 472 1015 682"> </div> <p> <math>(R_1 = -H, -CH_3, -C_2H_5, -C_3H_7, -C_4H_9</math>  <math>R_2 = -H, -CH_3, -C_2H_5, -C_3H_7, -C_4H_9</math>  <math>R_3 = -H, -CH_3, -C_2H_5, -C_3H_7, -C_4H_9</math>  <math>Y = BF_4, PF_6, ClO_4, F, Cl, Br, OH)</math> </p>
<p>10 Pyridinium salts</p>	<div data-bbox="706 1029 933 1228"> </div> <p> <math>(R = -H, -CH_3, -C_2H_5, -C_3H_7, -C_4H_9</math>  <math>Y = BF_4, PF_6, ClO_4, F, Cl, Br, OH)</math> </p>

Table 2: Solid Electrolytes

	Types	Concrete examples
5	Inorganic acid anion-alkali metal salts	$\text{XAsF}_6$ , $\text{XPF}_6$ , $\text{XBF}_4$ , $\text{XClO}_4$ (X=H, Li, K, Na)
	Organic acid anion-alkali metal salts	$\text{XCF}_3\text{SO}_3$ , $\text{XC}_n\text{F}_{2n+1}\text{SO}_3$ (n=2, 4, 8), $\text{XN}(\text{CF}_3\text{SO}_2)_2$ , $\text{XC}(\text{CF}_3\text{SO}_2)_3$ , $\text{XB}(\text{CH}_3)_4$ , $\text{XB}(\text{C}_6\text{H}_5)_4$ (X=H, Li, K, Na)
	Quaternary ammonium salts	$[\text{CH}_3(\text{CH}_2)_3]_4\text{N} \cdot \text{Y}$ , $\text{C}_n\text{H}_{2n+1}\text{N}(\text{CH}_3)_3 \cdot \text{Y}$ (n=10 - 18), $\text{C}_n\text{H}_{2n+1}\text{N}(\text{CH}_3)_2 \cdot \text{Y}$ (n=10 - 18), (Y= $\text{BF}_4$ , $\text{PF}_6$ , $\text{ClO}_4$ , F, Cl, Br, OH)
10	Anionic surfactants	$\text{C}_n\text{H}_{2n+1}\text{COO} \cdot \text{X}$ (n=10 - 18), $\text{C}_n\text{H}_{2n+1}\text{OC}_m\text{H}_{2m}\text{COO} \cdot \text{X}$ (n=10 - 18, m=10 - 18), $\text{C}_{10}\text{H}_7\text{COO} \cdot \text{X}$ , $\text{C}_n\text{H}_{2n+1}\text{C}_{10}\text{H}_6\text{COO} \cdot \text{X}$ (n=10 - 18), $\text{C}_n\text{H}_{2n+1}\text{SO}_3 \cdot \text{X}$ (n=10 - 18), $\text{C}_n\text{H}_{2n+1}\text{OC}_m\text{H}_{2m}\text{SO}_3 \cdot \text{X}$ (n=10 - 18, m=10 - 18), $\text{C}_{10}\text{H}_7\text{SO}_3 \cdot \text{X}$ , $\text{C}_n\text{H}_{2n+1}\text{C}_{10}\text{H}_6\text{SO}_3 \cdot \text{X}$ (n=10 - 18), $\text{C}_n\text{H}_{2n+1}\text{OSO}_3 \cdot \text{X}$ (n=10 - 18), (X=H, Li, K, Na)

[0077]

Organic solvents to be added are not limited specifically as long as it can compatible with each of the gelling agent and electrolyte.

[0078]

5

The stabilizer to be added to stabilize the self-

assembling compound is not specifically limited and may be selected from the known stabilizers compatible with the liquid electrolyte and the gelling agent in it.

[0079]

5 [EXAMPLE]

The present invention is described more specifically by Examples. The self-assembling compounds used were commercial products or synthesized by the known method.

10 [0080]

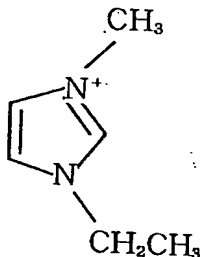
Example 1

Sample A, a compound of formula (1) where n is 2 and R is C<sub>9</sub> straight-chain alkyl, was tested for its gelating ability with imidazolium tetrafluoroborate (ImBF<sub>4</sub>) represented by formula (19), a liquid salt at room temperature, by the inverted test tube method.

[0081]

[Che38]

20



BF<sub>4</sub><sup>-</sup>

( 19 )

[0082]

25

ImBF<sub>4</sub> added with 3% of Sample A was put into a test tube, and heated at about 50°C or higher to dissolve, and then left standing to cool off at 25°C (room

temperature) for 1 hour, and then the condition was observed. Mixtures each added with 1% of an organic solvent were also tested. The results are given in Table 3.

5 [0083]

A mere mixture of  $\text{ImBF}_4$  and Sample A dissolved when heated, but not gelled with crystal precipitation. However, gelation occurred in the presence of a small amount of an organic solvent of various kind. When the gel thus formed was left standing at room temperature for 6 months, neither leakage of  $\text{ImBF}_4$  nor discoloration of the gel was observed.

[0084]

Table 3

15 ImBF <sub>4</sub> /Sample A mixture	Organic solvents added to the mixture				
	Toluene	THF + Acetonitrile	Ethanol	Methanol	DMF
x	○	○	○	○	○

(notes)

20 ○: The mixture was gelled.  
x: Crystal precipitation occurred, no gelation.

[0085]

#### Example 2

The same procedure as used in Example 1 was carried out except that a compound represented by formula (2) where n is 2 and R is  $\text{C}_{17}$  straight-chain alkyl (Sample B) was tested for its gelation ability with the room temperature liquid salt of Formula (19).



The mixture gelled without adding any organic solvent.

[0086]

When the gel was left standing for 6 months at room temperature, neither leakage of  $\text{ImBF}_4$  nor  
5        discoloration of the gel was observed. Further, addition of lithium tetrafluoroborate ( $\text{LiBF}_4$ ) as the solid electrolyte did not affect the gel.

[0087]

#### Example 3

10        The same procedure as in Example 1 was carried out except that Sample C, a compound represented by formula (3-1) where  $\text{R}^{10}$  is Z-L-valyl-L-valyl, was tested for its gelating ability with  $\text{ImBF}_4$ . The mixture gelled without  
15        adding any organic solvent. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0088]

[Che39]

20         $\text{R}^{10}\text{-NH-CH}_2(\text{CH}_2)_{16}\text{CH}_3$         (3-1)

[0089]

#### Example 4

The same procedure as in Example 1 was carried out except that an 1:1 mixture of Sample C, a compound  
25        represented by formula (4) where R is 3,7-dimethyloctyl, and Sample E, a compound represented by formula (5) where R is 3,7-dimethyloctyl, was analyzed

for its gelating ability with the liquid salt of formula (19). The results are given in Table 4.

[0090]

A mere mixture of  $\text{ImBF}_4$  and Samples D and E dissolved when heated, but not gelled with occurring of crystal precipitation. However, gelation occurred in the presence of a small amount of an organic solvent of various kind. When the gel thus formed was left standing at room temperature for 6 months, neither leakage of the solvent nor discoloration of the gel was observed.

[0091]

[Table 4]

Table 4

ImBF <sub>4</sub> only	Organic solvents added to the mixture		
	Toluene	THF + Acetonitrile	DMF
x	○	○	○

[0092]

#### Example 5

The same procedure as in Example 1 was carried out except that the compound represented by formula (6) where n is 2 and R is a straight-chain alkyl group having a carbon number of 17 (Sample F) was tested for its gelation capability with the liquid salt of formula (19). As a result, the mixture gelled without adding any organic solvent.

[0093]

When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed. Further, addition of lithium tetrafluoroborate ( $\text{LiBF}_4$ ) or lithium  
5 hexafluorophosphate ( $\text{LiPF}_6$ ) as the solid electrolyte did not affect the gel.

[0094]

#### Example 6

The same procedure as used in Example 5 was  
10 carried out except that  $\text{ImBF}_4$  was replaced by 1-butylpyridiumtetrafluoroborate represented by formula (20) below as the salt being liquid at room temperature, to test the gelation capability of the gelling agent. The mixture gelled without adding any  
15 organic solvent. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

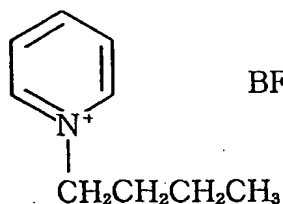
[0095]

Further, addition of lithium tetrafluoroborate  
20 ( $\text{LiBF}_4$ ) or lithium hexafluorophosphate ( $\text{LiPF}_6$ ) as the solid electrolyte did not affect the gel.

[0096]

[Che40]

25



( 20 )

[0097]

Example 7

The same procedure as used in Example 1 was carried out except that the derivative represented by  
5 formula (7) where R is C<sub>16</sub> straight-chain alkyl (Sample G) was tested for its gelation capability with the liquid salt of formula (19). The mixture gelled when a small amount of chloroform was added. When the gel was left standing for 6 months at room temperature, neither  
10 leakage of the solvent nor discoloration of the gel was observed.

[0098]

Example 8

The same procedure as used in Example 1 was  
15 carried out except that the derivative represented by formula (8) where R is C<sub>17</sub> straight-chain alkyl (Sample H) was tested for its gelation capability with the liquid salt of formula (19). The mixture gelled when a small amount of chloroform was added. When the gel was  
20 left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0099]

Example 9

25 The same procedure as used in Example 1 was carried out except that the derivative represented by formula (9) where R is C<sub>10</sub> straight-chain alkyl (Sample

I) was tested for its gelation capability with the liquid salt of formula (19). The mixture gelled without adding any organic solvent. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed. Further, addition of lithium tetrafluoroborate ( $\text{LiBF}_4$ ) or lithium hexafluorophosphate ( $\text{LiPF}_6$ ) as the solid electrolyte did not affect the gel.

[0100]

10 Example 10

The same procedure as used in Example 1 was carried out except that the derivative represented by formula (10) where  $\text{R}^5$  is a straight-chain alkyl group having a carbon number of 17 substituted with one hydroxyl group (12-hydroxystearic acid, Sample J) was tested for its gelation capability with the liquid salt of formula (19). The mixture gelled when chloroform was added. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0101]

Example 11

The same procedure as used in Example 1 was carried out except that the derivative represented by formula (11) where R is  $\text{C}_{11}$  straight-chain alkyl and  $\text{R}^1$  and  $\text{R}^2$  are butyl (N-launoyl-L-glutamic acid- $\alpha$ ,  $\gamma$ -bis-n-butylamide, Sample K) was tested for its gelation

capability with the liquid salt of formula (19). The mixture gelled when a small amount of chloroform was added. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor  
5    discoloration of the gel was observed.

[0102]

#### Example 12

The same procedure as used in Example 1 was carried out except that the derivative represented by  
10    formula (12) where  $R^1$  and  $R^2$  are isopropyl (Sample L) was tested for its gelation capability with the liquid salt of formula (19). The mixture gelled when a small amount of chloroform was added. When the gel was left standing for 6 months at room temperature, neither  
15    leakage of the solvent nor discoloration of the gel was observed.

[0103]

#### Example 13

The same procedure as used in Example 1 was  
20    carried out except that the derivative represented by formula (13) where  $R^1$  is 3,7-dimethyloctyloxycarbonylmethyl group and  $R^2$  is benzyl group [cyclo(L-asparagyl-L-phenylalanyl), Sample M] was tested for its gelation capability with the liquid salt  
25    of formula (19). The mixture gelled when a small amount of chloroform was added. When the gel was left standing for 6 months at room temperature, neither

leakage of the solvent nor discoloration of the gel was observed.

[0104]

Example 14

5           The same procedure as used in Example 1 was carried out except that the derivative represented by formula (14) (Sample N) was tested for its gelation capability with the liquid salt of formula (19). The mixture was gelled when chloroform was added. When the  
10   gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0105]

Example 15

15           The same procedure as used in Example 1 was carried out except that the derivative represented by formula (15) where R is a decyl group having a carbon number of 10 (Sample O) was tested for its gelation capability with the liquid salt of formula (19). The  
20   mixture gelled when chloroform was added. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0106]

25   Example 16

          The same procedure as used in Example 1 was carried out except that the derivative represented by

formula (16) (Sample P) was tested for its gelation capability with the liquid salt of formula (19). The mixture gelled when chloroform was added. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0107]

#### Example 17

The same procedure as used in Example 1 was carried out except that the compound represented by formula (17) where q is 5, m is 18 and R<sup>9</sup> is isopropyl (Sample Q) was tested for its gelation capability with the liquid salt of formula (19). The mixture gelled without adding any organic solvent. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed. Further, addition of lithium tetrafluoroborate (LiBF<sub>4</sub>) or lithium hexafluorophosphate (LiPF<sub>6</sub>) as the solid electrolyte did not affect the gel.

[0108]

#### Example 18

The same procedure as used in Example 17 was carried out except that 1-butylpyridiumtetrafluoroborate (PyBF<sub>4</sub>) represented by formula (20) was used as the liquid salt at room temperature, to test gelation capability of the gelling agent. The mixture gelled without adding any organic



solvent. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0109]

- 5           Further, addition of lithium tetrafluoroborate ( $\text{LiBF}_4$ ) or lithium hexafluorophosphate ( $\text{LiPF}_6$ ) as a solid electrolyte did not affect the gel.

[0110]

#### Example 19

- 10           The same procedure as used in Example 1 was carried out except that the compound represented by formula (18) where both  $\text{R}^6$  and  $\text{R}^7$  are methyl group (Sample R) was used as the gelling agent for the liquid salt of formula (19), and potassium sorbate was added  
15 as the stabilizer. The mixture gelled without adding any organic solvent. When the gel was left standing for 6 months at room temperature, neither leakage of the solvent nor discoloration of the gel was observed.

[0111]

- 20           Comparative Example 1

- Polymethyl methacrylate having a molecular weight of 7,000 was added to the liquid salt of formula (19) and the mixture was heated at  $90^\circ\text{C}$  and cooled to test its gelation capability. When the added amount of  
25 polymethylmethacrylate was 20% or less, the mixture did not gelate, showing leakage of the solution.

[0112]

[Effect of the Invention]

As described above, the present invention provides a gel electrolyte serviceable for a long period of time, superior in mechanical strength and  
5 electroconductivity.

The gel electrolyte of the present invention can be produced by a much simpler process with a smaller number of steps than the conventional processes for the polymer gel electrolyte production with less production  
10 cost, because it can be prepared only by heating a mixture of the gelling agent and liquid electrolyte to accelerate dissolution, followed by cooling.

[Name of the Document] ABSTRACT OF THE DISCLOSURE

[ABSTRACT]

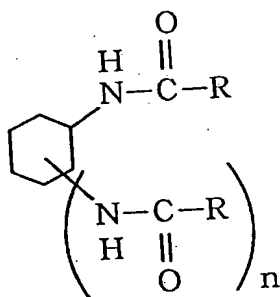
[Object] The object is to provide a gel electrolyte being stable for a long period of time without

5    discoloring, which can gelate with a small amount of a gelling agent with reduction in electroconductivity as small as possible.

[Means for Achievement]    A gel electrolyte containing at least a gelling agent represented by formula (1) and  
10    a material of high ion conductivity which is liquid at working temperature.

[Che 1]

15



( 1 )

( n = 0, 1, 2 )

20

wherein R is hydrogen, a straight or branched aliphatic hydrocarbon group of 1-29 carbon atoms.

[Elected Figure]    None

10-313938

[Name of the Document]	Authorized Correction Data
[Document to be corrected]	Patent Application
<Recognition Information Additional Information>	
[Application]	offeror
[Identification No.]	000001007
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(Reason for Change) New Registration

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